Comment on "Fermi Surface, Surface States, and Surface Reconstruction in Sr_2RuO_4"

In a recent Letter, Damascelli *et al.* [1] reported angle-resolved photoemission spectra on Sr_2RuO_4 with the aim of elucidating the electronic structure in the vicinity of the Fermi energy. They interpret their data in terms of a surface state just below E_F near the M point, shadow bands induced by the $\sqrt{2} \times \sqrt{2}$ surface reconstruction, and Ru $4d$ t_{2g} bands that are consistent with de Haas–van Alphen (dHvA) measurements [2]. Although these results resolve the discrepancy which existed between earlier photoemission spectra and the Fermi surface topology derived from dHvA data, we point out that the bulk band structure of Sr_2RuO_4 [3] provides no evidence of a band gap supporting the existence of a surface state. Thus the interpretation given in [1] of the prominent emission feature near M in terms of a surface state localized in k space cannot be correct. A clarification of the origin of this feature, however, is crucial since it sheds light on the usefulness of angle-resolved photoemission as a probe of Fermi surface electronic properties and since the controversial t_{2g} band in Sr_2RuO_4 shares important characteristics with the key electronic band in the high-T_c superconductors. Also, a clear picture of the electronic properties of Sr_2RuO_4 is desirable in view of the fact that this system is the only layered perovskite without copper that exhibits superconductivity [4] and appears to show unconventional spin-triplet pairing with a p-wave order parameter [5].

An interpretation of the observed photoemission spectra that is consistent with all available experimental and theoretical results can be achieved by associating the feature near M with emission from the deformed xy band within the reconstructed first layer of Sr_2RuO_4. According to recent surface electronic structure calculations, scanning tunneling microscopy and low energy electron diffraction data [6], the following picture emerges for the properties of the first layer: The reconstruction is driven by the freezing of a zone boundary soft phonon mode giving a slight rotation of the RuO_6 octahedra around the surface normal. The rotation reduces the effective d-d hopping within the first Ru plane and causes band narrowing. In the nonmagnetic case (which seems to be observed in the photoemission work) the band narrowing yields a shift of the xy Van Hove singularity (VHs) below E_F, giving rise to strong emission near M.

The photoemission spectra in this picture consist of a superposition of two sets of t_{2g} bands originating in the surface layer and the deeper layers. Both sets are energetically very similar and difficult to resolve, with the exception of the xy VHs near M: in the bulk it is above E_F while at the surface it is below. Accordingly, the γ sheet of the Fermi surface is electronlike in the bulk, but holelike at the surface. Whereas the bulk xy band crosses the Fermi level along ΓM, the reconstruction-induced xy band remains below E_F near M and crosses along MX. Both bands cross the Fermi level along ΓX. In addition, the reconstruction generates weak shadow bands, as observed in [1].

The weak momentum dispersion of the xy band near M can be understood in terms of Coulomb correlations between Ru d electrons. As shown recently using perturbation theory and quantum Monte Carlo calculations for a multiband Hamiltonian [7], an on-site Coulomb energy $U \approx 1$ eV leads to a quasiparticle self-energy near E_F of the same order as the binding energy, giving rise to considerable flattening of the momentum dispersion and strong effective mass enhancement. Correlations also cause a slight charge transfer from the narrow xz, yz bands to the wider xy band, strong quasiparticle damping, and an overall narrowing of the xz, yz bands by about a factor of 2, in agreement with photoemission data. None of these aspects can be understood within the single-particle picture and they demonstrate the importance of considering correlation effects when interpreting photoemission spectra.

The sensitivity of the xy band to surface degradation is at first sight surprising since adsorption of atoms or molecules most likely involves primarily xz, yz states rather than planar xy states. Nevertheless, adsorption requires reorganization of all Sr $6s$, O $2p$, and Ru $4d$ electrons in the first layer. It is plausible that this affects also the VHs near M. Quantitative chemisorption calculations might help to clarify this point.

In conclusion, the Sr_2RuO_4 photoemission spectra of Damascelli *et al.* [1] are interpreted in terms of a superposition of bulk t_{2g} states consistent with dHvA measurements and deformed t_{2g} states existing in the reconstructed first layer. The main deformation arises near M where the VHs of the xy band is below E_F as a result of a slight narrowing of the t_{2g} bands, while the bulk VHs is unoccupied.

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